

### 3.0 RECYCLED URANIUM MASS FLOW

#### 3.1 URANIUM RECYCLE DESCRIPTION

For purposes of this project, RU has been defined as any uranium that has been irradiated in a reactor and as a result contains TRU (e.g., Pu and Np) and fission products (e.g.,  $^{99}\text{Tc}$ ). The methodology applied in this project for identifying ORGDP's involvement with the flow of RU materials involves: (1) the source site and (2) the  $^{235}\text{U}$  assay of the material. Sites identified as RU candidate source sites are the U.S. government facilities at Hanford and Savannah River that operated production reactors and used chemical separation processes to extract uranium from irradiated fuel, Harshaw Chemical Company, and foreign customers for U.S enrichment services. Secondary sites providing RU to ORGDP included PGDP, PORTS, and ORNL.

Data for ORGDP transactions with these RU candidate source sites was extracted from Material Balance Reports (MBR) issued by the site Nuclear Material Control and Accountability (NMC&A) organization. These reports provide official accountability data for all site uranium and for other accountable nuclear materials, including the name or symbol-code of the accountability station with which the receipt from or shipment to has occurred, material type, amount of uranium,  $^{235}\text{U}$ , and the  $^{235}\text{U}$  assay. The earlier MBRs listed the name and location of the accountability station (e.g., General Electric Company, Richland, Washington) rather than the accountability station symbol-code (e.g., HGE). The accountability station symbol-code, now referred to as the Reporting Identification Symbol (RIS), began appearing on the ORGDP MBRs in FY 1964.

The second level for identification of RU involves the  $^{235}\text{U}$  assay of the uranium. Based on process knowledge relative to assay ranges, the annual average assay of RU receipts from the source sites provides a basis for tracking RU in inventories, feed to the gaseous diffusion process, and shipments.

Under the NMC&A program, uranium is an accountable nuclear material. However, RU is not separately accountable. It should be recognized that the methodology utilized in this project for identifying and tracking RU is imperfect, and some level of RU accountability is unavoidably lost. Physical losses of RU occurred which could not be identified and quantified. Loss of accountability likely occurred as a result of assay blending of RU with non-RU with the result that the RU could no longer be tracked. Other losses of accountability may have occurred as a result of data unavailability or the misinterpretation of data. Losses are discussed further in Section 3.4.

#### 3.2 URANIUM RECEIPTS

ORGDP first received RU from Hanford in FY 1952 when 99,970 kg of  $\text{UO}_3$  was recorded. Receipts from Hanford continued from FY 1952 through FY 1962. During the period 1952 through 1958, the annual average  $^{235}\text{U}$  assay range for Hanford RU receipts was 0.646% to 0.666%. Beginning in FY 1959, the assay changed from depleted to slightly enriched in the range of 0.848% to 0.864%. In total, 4,276,111 kgU of RU was received from Hanford. Annual receipts are summarized in Table 3.2-1.

**Table 3.2-1. RU Received at ORGDP from Primary Source Sites**

Fiscal Year	Hanford		Harshaw		Savannah River		Foreign		Total kgU
	kgU	Assay	kgU	Assay	kgU	Assay	kgU	Assay	
1952	99,970	0.646							99,970
1953	578,249	0.666	1,402,761	0.666					1,981,010
1954	1,115,345	0.666	299,574	0.671					1,414,919
1955	526,475	0.657			271,949	0.682			798,424
1956	323,882	0.665			2,538,844	0.670			2,862,726
1957	98,218	0.652			2,635,163	0.667			2,733,381
1958	7,201	0.649			1,077,065	0.648			1,084,266
1959	261,253	0.848			828,250	0.625			1,089,503
1960	609,775	0.856			1,677,456	0.603			2,287,231
1961	611,020	0.853			1,121,645	0.598			1,732,665
1962	44,722	0.864			139,308	0.590			184,030
1963	1	0.650							1
1964 - 1968									
1969							2,033	1.332	2,033
1970							20,532	1.724	20,532
1971							4,734	1.698	4,734
1972							24	2.151	24
1973							61,531	1.638	61,531
1974							115,373	0.989	115,373
1975							73,892	0.888	73,892
1976+TQ							86,145	0.746	86,145
1977							55,965	1.090	55,965
1978							28,355	1.193	28,355
1979							46,454	1.037	46,454
1980							88,047	1.262	88,047
1981							67,078	1.011	67,078
1982									
1983							257,687	1.525	257,687
1984							173,916	1.257	173,916
1985									
1986							211,140	0.947	211,140
1987							1	3.207	1
1988							1,451	1.118	1,451
1989 - 1999									
TOTALS*	4,276,111		1,702,335		10,289,680		1,294,359		17,562,485

\* Numbers may not sum because of rounding.

During FY 1953 and FY 1954, 1,702,335 kgU of RU was delivered to ORGDP from Harshaw Chemical Company. This material had previously been delivered to Harshaw from Hanford. Documentation found in *AEC Accountability Survey Reports, Reports for Period October 1947 Through May 27, 1953 (U)*<sup>1</sup> states that:

The feed manufacture plant began processing depleted uranium from the Hanford recovery process in June 1952. Difficulties, attributed to impurities in the recovered oxide, were experienced in processing this material. Consequently, during September the feed manufacture plant resumed operations in normal uranium trioxide from Harshaw and uranium tetrafluoride from Mallinckrodt. Present plans are to remove the objectionable impurities and render the Hanford recovered material chemically more reactive at Harshaw prior to processing it in the feed plant at Carbide K-25.

Receipts of RU from Savannah River were first recorded in FY 1955 and continued through FY 1962. During this time, 10,289,680 kgU was received. The <sup>235</sup>U annual average assay range for receipts of Savannah River RU was 0.590% to 0.682%. Based on process

<sup>1</sup> *AEC Accountability Survey Reports, Reports for Period October 1947 Through May 27, 1953 (U)*

knowledge relative to assay for reactor fuels used at Hanford and Savannah River and on receipts from both, assay ranges for tracking Hanford and Savannah River RU at ORGDP were established as 0.59% to 0.69% for depleted and 0.84% to 0.87% for enriched. Inventories and transactions on the outer boundaries of these assay ranges were considered on a case-by-case basis.

Beginning in FY 1969, RU was received at ORGDP from foreign sources primarily in the form of UF<sub>6</sub>. All foreign receipts, excluding normal assay assumed to be non-RU, were analyzed. Non-normal receipts from foreign entities continued through FY 1988, with the exception of FY 1982 and FY 1985 when no foreign receipts were recorded. All receipts ranged in assay from 0.746% <sup>235</sup>U to 3.2% <sup>235</sup>U.

Data collected from ORGDP MBRs indicates that from FY 1969 through FY 1988, a total of 1,294 MTU of non-normal material was received from foreign sources. Table 3.2-2 provides a summary of receipts by country.

**Table 3-2.2. Foreign Reactor RU Returns to ORGDP**

Country	MTU
France	843
United Kingdom	296
Germany	132
Netherlands	10
International Account*	10
Belgium	2
Japan, Namibia, India, and Sweden	1
Total	1,294

\*From material distributed by the Division of International Affairs.

Receipts of 843 MTU from France at assays of less than 2% are believed to be RU. It is known that the French had the reprocessing capability. The United Kingdom returned 296 MTU. The United Kingdom material is believed to be RU, and 231 MTU of this material was fed to the cascade in FY 1975, FY 1976, the FY 1976 transition quarter, and FY 1978. It appears that 12 MTU of French material was fed in FY 1978. Quantities and assay range of foreign receipts are shown in Table 3-2.3.

**Table 3-2.3. Quantities of Assay Range of Foreign Reactor Returns**

Assay Range % <sup>235</sup> U	Quantity (MTU)	Average Assay % <sup>235</sup> U	Percent of Total
≤ 0.69	243	0.643	18.8
0.715 - 1.5	810	0.979	62.6
1.51 - 2.5	151	2.048	11.7
≥ 2.51	90	2.956	6.9
TOTAL	1,294		

In FY 1986, a year after ORGDP was placed in standby, 486 MTU of foreign uranium at average assays of about 1% was shipped to PGDP. PGDP recorded this as receipts of RU. One cylinder of approximately 1 MTU was returned to France in FY 1988. This material plus the 243 MTU depleted that is known to have been fed to the cascade leaves about 565 MTU to be accounted for. Of the amount of RU shipped to PGDP, 2,810 kgU was categorized by ORGDP as UF<sub>6</sub> heels. It is assumed that the full content of these cylinders, that now contain only heels, was fed to the ORGDP cascade and thus accounts for the 565 MTU. The 2,810 kgU heels represents about 367 30B cylinders, assuming a maximum heel of 11.34 kg UF<sub>6</sub> (7.666 kgU). The maximum full shipping weight limit per cylinder is 2,277 kg UF<sub>6</sub> (1,539 kgU). Therefore, the 367 cylinders can account for the 565 MTU as likely being fed to the ORGDP cascade.

In addition to receipts from source sites, ORGDP received material identified as RU from PGDP, PORTS, and ORNL. These receipts were identified from the MBRs based primarily on defined assay ranges and are presented on Table 3.2-4. The PGDP and PORTS RU was primarily in the form of UF<sub>6</sub>. Receipts from ORNL were in the form of UO<sub>3</sub> and UF<sub>4</sub>.

**Table 3.2-4. RU Received at ORGDP from Secondary Sites**

Fiscal Year	PGDP		PORTS		ORNL		TOTAL kgU
	kgU	Assay	kgU	Assay	kgU	Assay	
1952					12	0.656	12
1953	153,111	0.637					153,111
1954	21,396	0.669			694	0.653	22,090
1955	33,426	0.655			1,134	0.661	34,560
1956	27,341	0.669	3,048	0.673	830	0.655	31,219
1957	34,906	0.661			2,586	0.653	37,492
1958	29,020	0.670			4	0.670	29,024
1959	70,151	0.652			1	0.593	70,152
1960	2,091	0.642					2,091
1961	244	0.634			12	0.671	256
1962	10,511	0.634					10,511
1963	35,489	0.640					35,489
1964	9,052	0.641					9,052
1965	464	0.640					464
1966 - 1968							0
1969	236,325	0.643					236,325
1970	420,388	0.656			1	0.630	420,389
1971 - 1999							
TOTALS*	1,083,914		3,048		5,274		1,092,236

\* Numbers may not sum because of rounding.

### 3.3 URANIUM SHIPMENTS

Shipments of RU from the ORGDP site were identified from the MBRs based on the assay ranges for RU receipts as discussed in Section 3.2. ORGDP shipments are summarized in Table 3.3-1.

Shipments to PGDP are presented by fiscal year in Table 3.3-2. Shipments to PGDP within the assay ranges defined as RU as shown in the MBRs were 13,994,541 kgU. This amount has been reduced by 1,946,116 kgU withdrawn from the ORGDP cascade and 419,096 kgU received from the Y-12 Plant within the defined assay range for RU, both of which were determined to be non-RU. Based on timing of these receipts of non-RU and shipments to PGDP,

it was determined that the most likely disposition of the non-RU was to PGDP. However, it is recognized that some part of this non-RU could have been fed to ORGDP.

**Table 3.3-1. ORGDP Shipments of RU**

Receiving Site	kgU
PGDP	11,629,329
PORTS	301,077
Y-12 Plant	189,146
ORNL	7,589
Savannah River	11,057
Fernald	1,909
Foreign	1,451
<b>TOTALS</b>	<b>12,141,558</b>

**Table 3.3-2. ORGDP RU Shipments to PGDP**

Fiscal Year	MBR Total Assay Range Shipments (kgU)	Adjustments		Net Shipments (kgU)
		Cascade Withdrawals (kgU)	Receipts from Y-12 (kgU)	
1952	7		1,310	0
1953	2,738,891	976,490	2,601	1,758,490
1954	1,768,711	120,163	139,919	1,508,629
1955	1,403,643	452,068	29,153	922,422
1956	2,669,834		112,659	2,557,175
1957	3,181,798		31,700	3,150,098
1958	29,096		29,033	63
1959	514,635		43,551	471,084
1960	580,679		28,468	552,211
1961	81,039		702	80,337
1962	46,825			46,825
1963	64,711			64,711
1964	17,498			17,498
1965 - 1969				
1970	14,126			14,126
1971				
1972	397,395	397,395		0
1973				
1974	(3)			(3)
1975 - 1985				
1986	485,656			485,656
1987 - 1999				
<b>TOTALS*</b>	<b>13,994,541</b>	<b>1,946,116</b>	<b>419,096</b>	<b>11,629,322</b>

\* Numbers may not sum because of rounding.

In addition to off-site shipments, 5,914,681 kgU of RU, including 807,172 kgU of foreign receipts, were fed to the ORGDP cascade. Feed to the ORGDP cascade is presented in Table 3.3-3.

**Table 3.3-3. Summary of Feed to ORGDP Based on Cumulative Cascade MBRs**

Year		Reactor Returns (MTU)	Normal (MTU)	PGDP Product (MTU)	Other (Inc. Refeed) (MTU)	Total Feed (MTU)
CY	1947		592		111	703
CY	1948		674		101	775
CY	1949		674		511	1,185
CY	1950		822		462	1,284
JAN - JUN	1951		456		669	1,125
FY	1952		1,299		5,761	7,060
FY	1953	153	1,100	1,664	5,307	8,224
FY	1954		1	3,591	20	3,612
FY	1955	3	5	3,703	110	3,821
FY	1956		264	4,149	39	4,452
FY	1957			4,604	32	4,636
FY	1958		116	3,380	1,482	4,978
FY	1959	660	1,398	3,292	9	5,359
FY	1960	1,949	876	2,930	20	5,775
FY	1961	1,259	1,947	2,933	3	6,142
FY	1962	424	2,408	2,851	23	5,706
FY	1963	5	2,109	2,871	47	5,032
FY	1964	4	2,654	2,184	483	5,325
FY	1965			2,126	5,053	7,179
FY	1966	4	2	2,112	5,669	7,787
FY	1967			1,931	5,497	7,428
FY	1968			1,730	5,062	6,792
FY	1969	2	1,521	2,713	402	4,638
FY	1970	377	1,811	2,637	43	4,868
FY	1971	5	2,918	2,832	577	6,332
FY	1972		1,542	2,782	367	4,691
FY	1973	62	3,557	1,875	147	5,641
FY	1974	287	3,723	2,060	0	6,070
FY	1975	91	4,454	1,891	0	6,436
FY	1976	70	4,000	2,050	15	6,135
JUL - SEP	1976	74	956	412	1	1,443
FY	1977	60	4,264	1,954	0	6,278
FY	1978	66	4,929	1,131	151	6,277
FY	1979	32	4,847	1,218	394	6,491
FY	1980	31	4,156	2,099	367	6,653
FY	1981	67	7,271	4,945	1,260	13,543
FY	1982		3,444	3,457	123	7,024
FY	1983	150	3,368	2,689	310	6,517
FY	1984	79	1,493	1,294	154	3,020
FY	1985		2,361	2,295	602	5,258
TOTALS*		5,915	78,012	86,385	41,384	211,695

\* Numbers may not sum because of rounding.

Reactor returns listed on Table 3.3-3 are based on assays received and fed in the ranges of 0.59% - 0.69% and 0.848% - 0.864%. For purposes of this table, normal uranium includes all feed to the cascade in the assay range of 0.70% - 0.72%. PGDP product includes all enriched

feeds which can be identified with receipts from PGDP. Some judgement was required for identifying the PGDP product since annual feed quantities include a blending of assays. Other feed recorded on Table 3.3-3 includes refeed of uranium previously withdrawn as tails and other miscellaneous feeds. Data for FY 1982 and FY 1985 were collected from NMMSS Reports versus MBRs.

### 3.4 RECYCLED URANIUM WASTE

Accountability data for uranium as reported in the MBRs does not identify losses at a level that can be associated specifically with RU. Cumulative losses and RU material unaccounted for (MUF) are calculated and presented in the ORGDP RU Mass Balance Summary, Table 3.4-1, as 598,192 kgU, or approximately 3% of total RU receipts. The project team was informed by individuals who were familiar with commercial uranium operations similar to the ORGDP feed plant—but who were using more recent technologies—that standards for normal operating losses are approximately 0.5%.

**Table 3.4-1. ORGDP RU Mass Balance by Fiscal Year**

Fiscal Year	Total Receipts (kgU)	Shipments							Fed to GDP (kgU)	Cumulative Losses and MUF (kgU)	Ending Inventory (kgU)
		PGDP (kgU)	PORTS (kgU)	Y-12 Plant (kgU)	ORNL (kgU)	Savannah River (kgU)	Fernald (kgU)	Foreign (kgU)			
1952	99,982	0		1,381	20						98,581
1953	2,134,121	1,758,497		2,370	121				153,000		318,714
1954	1,437,009	1,508,629		143,192	2,447		1,429				100,026
1955	832,984	922,422		14,563	3,635	8			3,000		(10,618)
1956	2,893,945	2,557,175	296,327	22,883	293	2					6,647
1957	2,770,873	3,150,098	3,322	2,655	1,062	4,752					(384,369)
1958	1,113,290	63			1	1,519					727,338
1959	1,159,655	471,084		11	2	1,403			660,000		754,493
1960	2,289,322	552,211		2,091	8	2,026			1,949,000		538,479
1961	1,732,921	80,337	1,427			1,347	208		1,259,000		929,081
1962	194,541	46,825							424,000		652,525
1963	35,490	64,711	1						5,000		618,303
1964	9,052	17,498							4,000		605,857
1965	464										606,321
1966									4,000		602,321
1967 - 1968											602,321
1969	238,358								2,033		838,646
1970	440,921	14,126							376,532		888,909
1971	4,734								4,734		888,909
1972	24								24		888,909
1973	61,531								61,531		888,909
1974	115,373	(3)							287,000		717,285
1975	73,892								91,039		700,138
1976	86,145								144,297		641,986
1977	55,965								60,362		637,589
1978	28,355								66,355		599,589
1979	46,454								31,785		614,258
1980	88,047								31,081		671,224
1981	67,078								67,078		671,224
1982									150,595		520,629
1983	257,687								79,235		699,081
1984	173,916										872,997
1985											872,997
1986	211,140	485,656									598,481
1987	1										598,482
1988	1,451							1,451			598,482
1989										598,482	
1990 - 1999											
TOTALS*	18,654,721	11,629,329	301,077	189,146	7,589	11,057	1,909	1,451	5,914,681	598,482	

\* Numbers may not sum because of rounding.

**NOTE:** Negative RU inventories in 1955 and 1957 result from the inability to accurately match by year gross shipments to PGDP with credits for cascade withdrawals (non-RU) within the same assay range as RU. Shipments to PGDP overall have been reconciled with PGDP receipt data.

The ORGDP feed plant, which began processing RU in 1952, represented a new technology and might be expected to have experienced greater process losses than more recent technologies. The feed plant process equipment was decontaminated and maintenance performed in Building K-1410. RU fluorination tower ash contained appreciable uranium as

well as TRU and various fission products. Some of this uranium was recovered while the rest was shipped to Paducah. The feed plant experienced many operating problems resulting in unmeasured releases of  $\text{UF}_6$  to the atmosphere, loss of uranium as  $\text{UO}_3$  and  $\text{UF}_4$  to the environment, and the discard of wash solutions from K-1410 to Poplar Creek. Residual uranium “heels” amounting to several hundred pounds were left in the  $\text{UF}_6$  feed cylinders. Interviews with former ORGDP personnel revealed that feed cylinders were not always exclusively reused as feed cylinders. There is some potential that feed cylinders, heels included, could have been used for tails withdrawal, hence the possibility that depleted uranium tails cylinders, now in storage at ORGDP, PGDP and PORTS may still contain these heels. There is little indication in the historical records that this material was recovered. Some of it was likely buried in the K-33 burial ground as low level waste. The balance was probably shipped to Paducah. Cascade compressors, converters, and other enrichment components containing RU deposits were decontaminated in Buildings K-1303 and K-1420. The wastewater generated in K-1303 was generally discharged to the K-1407B holding pond with little pre-treatment. The wastewater from K-1420, on the other hand, was processed for uranium recovery and then discharged to the K-1407B pond. Uranium recovery from these various maintenance facilities was not quantitative until after K-1420 was placed into operation, and even then recovered RU has lost its identity in the historical records. Significant uranium losses also occurred through the various purge cascade process vents associated with the enrichment plant. A process loss of 1 to 2% may be more realistic for the ORGDP feed plant. The additional 1 to 2% shown here likely results in part from the loss of accountability previously described.

### **3.5 RECYCLED URANIUM SCRAP**

RU scrap can be identified primarily in two areas. Ash generated in the feed plant fluorination tower was pulverized and recycled through the top of the tower. When it was no longer practical to recover uranium through the ORGDP process, the remaining ash was shipped off-site (primarily to PGDP) for further processing and disposal. In addition, uranium holdup in process equipment, filters, and containers was processed, and the uranium recovered. Accountability for the uranium as RU was lost when the RU went through the recovery process.

### **3.6 INVENTORY AS OF MARCH 31, 1999**

All RU received at the ORGDP site had been either shipped off-site or fed to the cascade as of March 31, 1999. Table 3.4-1 summarizes RU activity at ORGDP.